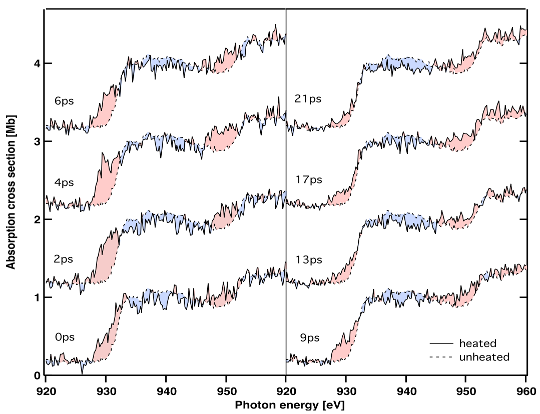
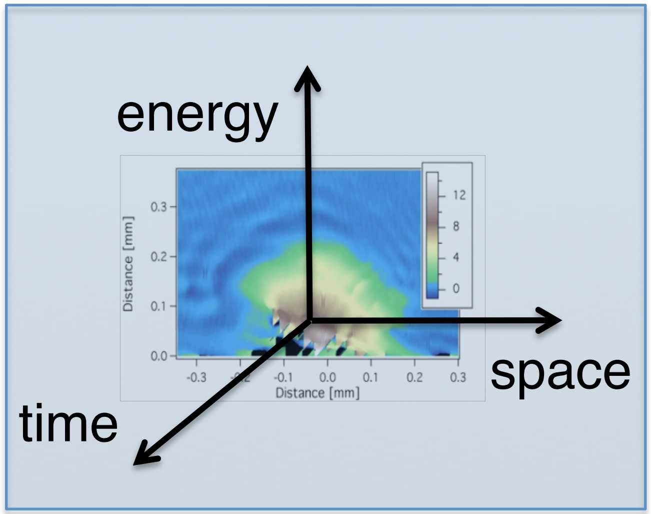
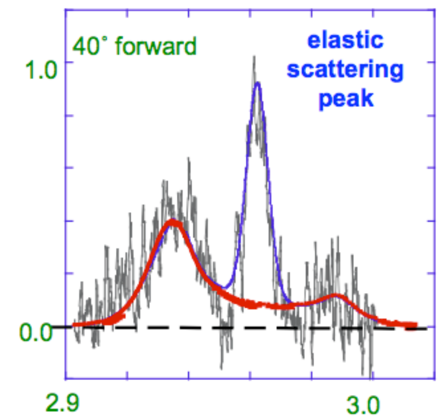
X-Ray Probes
of Warm Dense Matter and Hot Dense Matter

absorption

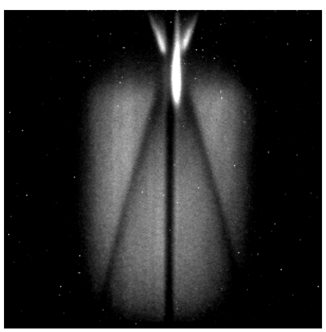




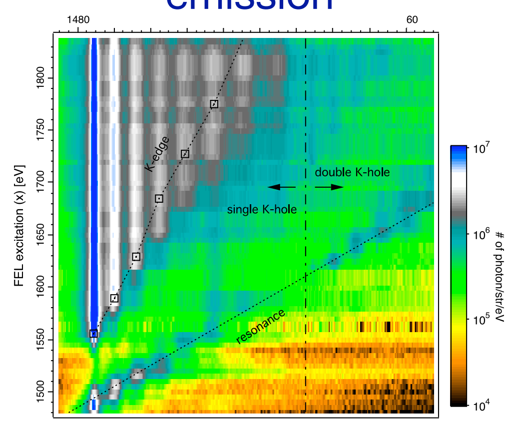
scattering



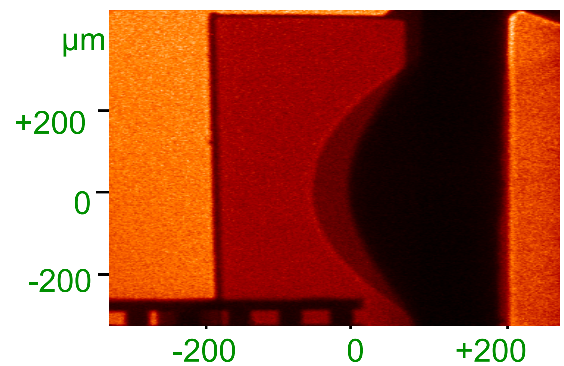
radiography



emission



imaging and diffraction

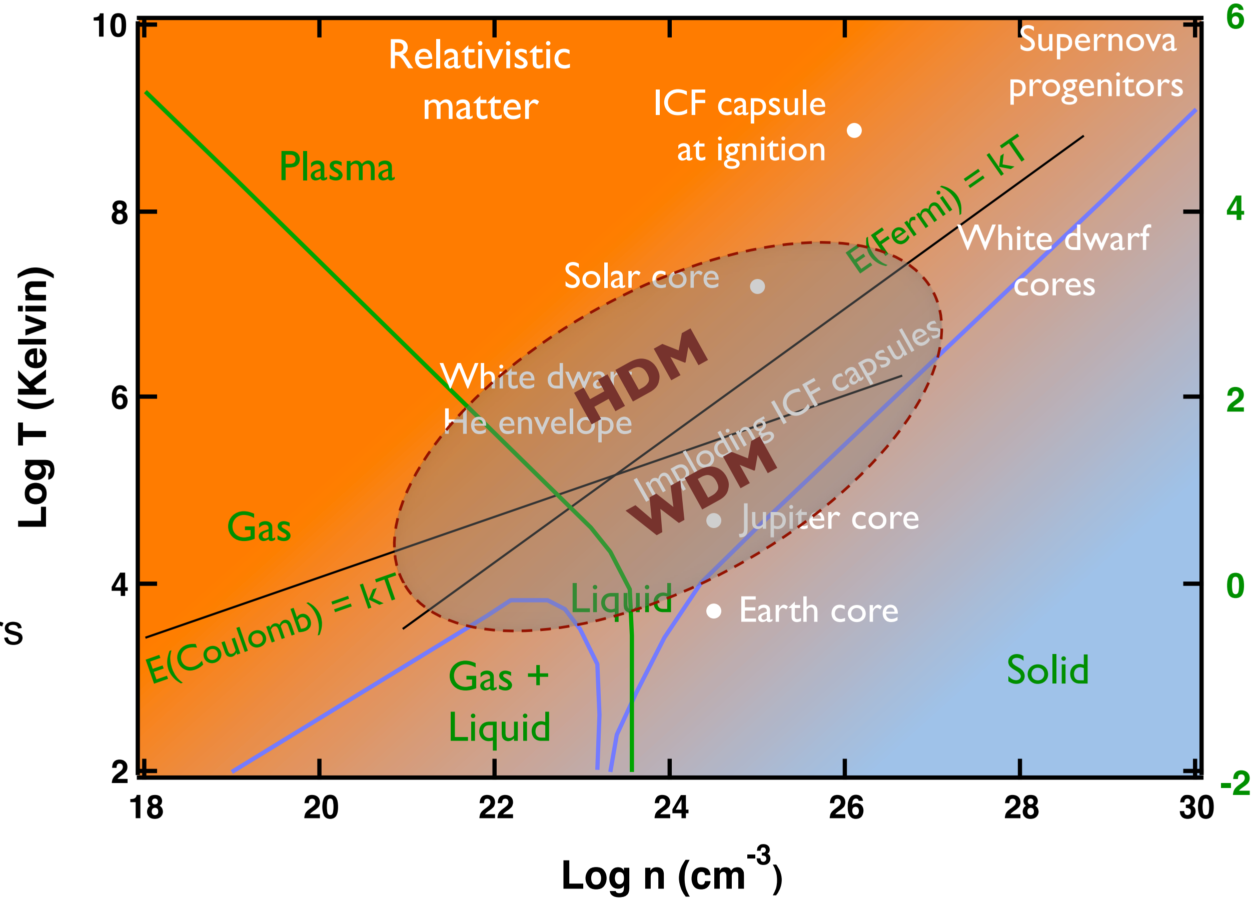


Roger Falcone (Physics, UCB and ALS, LBNL)
Phil Heimann (SLAC), Kyle Engelhorn (UCB), Byoung-ick Cho (Gwangju)

Interplay of physical processes in warm and hot dense matter (WDM and HDM) creates an interesting phase space for extreme conditions

Warm Dense Matter (WDM)
- Cores of large planets
- Shocked materials
- Materials in extreme environments
- Inertial confinement fusion (early time)

Hot Dense Matter (HDM)
- Stellar and supernova interiors
- Plasma devices: laser heated plasmas and Z-pinches
- Inertial confinement fusion (late time)



Basic Research Needs for High Energy Density Laboratory Physics, DOE Office of Science and NNSA (2010)

The Berkeley campus program (funding - SSAA to Joint Program) utilizes tools in Bay Area for studies of WDM, HDM, and plasmas

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LCLS provides x-ray pulses with sufficient energy for isochoric heating of matter, with sufficient photon energy and flux for inelastic scattering measurements of electron & ion temperature & densities, as well as high peak power and high energy optical lasers, to drive plasmas and shock compressed matter.

Time dependent x-ray techniques can uniquely probe matter under extreme and dynamic conditions that change rapidly

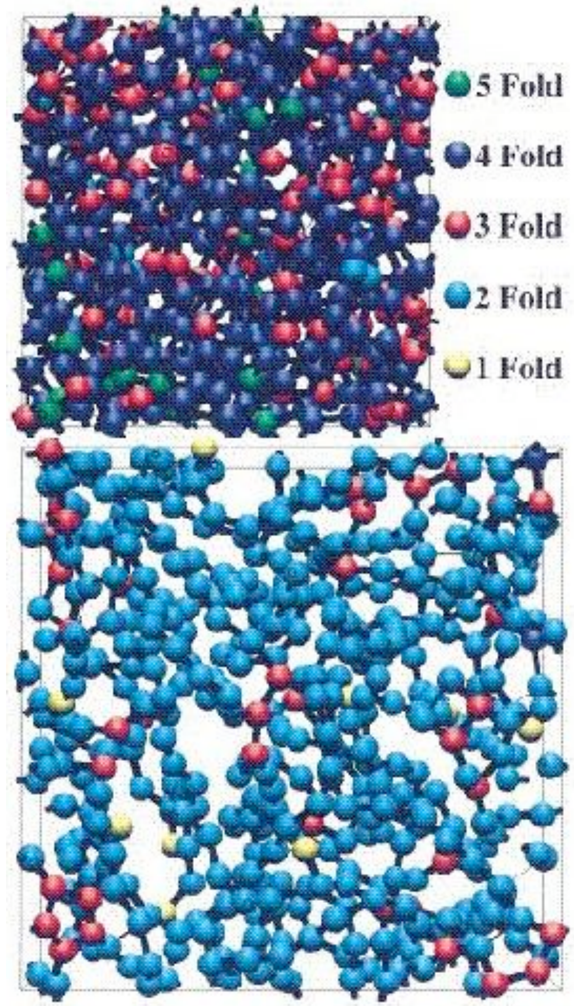
(1) Near edge absorption spectroscopy reveals electronic structure, bonding, nearest neighbors, and local order; radiography yields line density

(2) Diffractive scattering and imaging reveals structural on the nanoscale

(3) Inelastic scattering reveals electron and ion temperatures as well as plasma densities

(4) Emission spectroscopy reveals pathways for production of highly excited states in strongly-driven, dense systems

(5) Ionization mass spectroscopy reveals mass products of evolving, excited molecular systems



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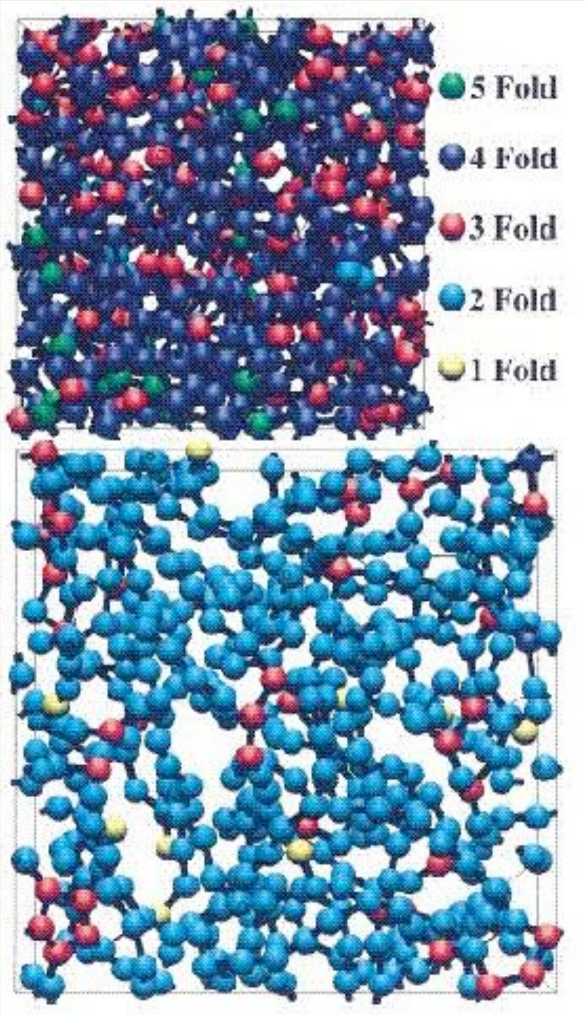
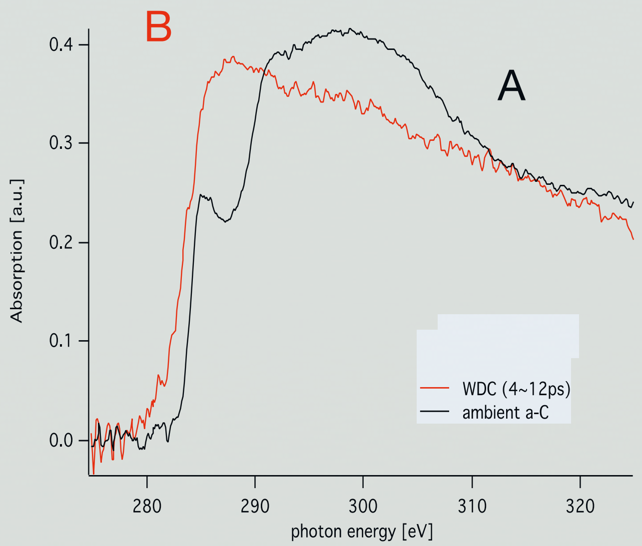
Can we validate simulations of warm dense matter?

A

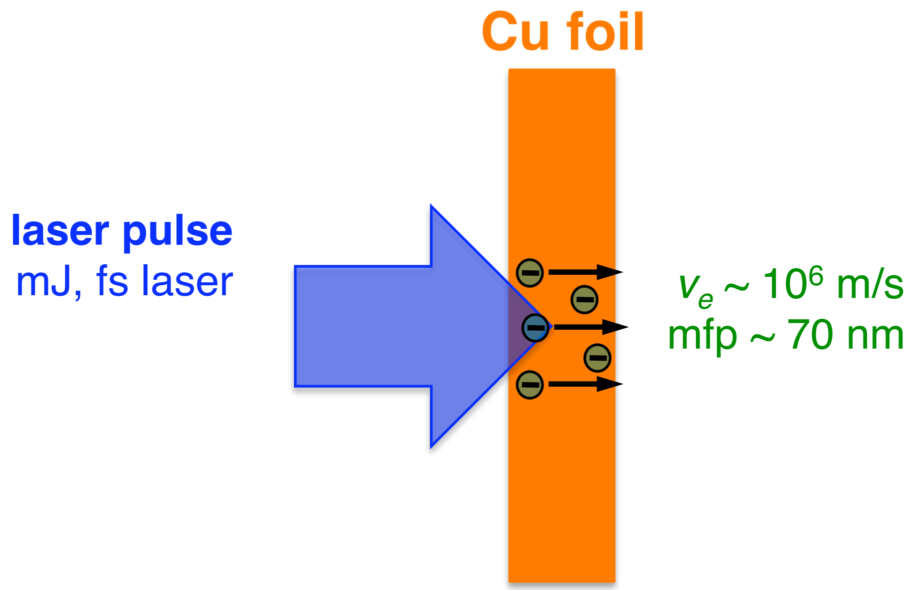
B

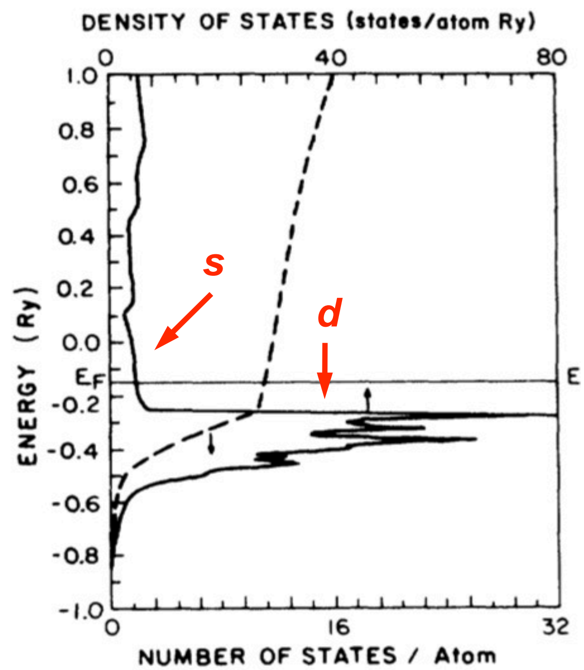




Structure and electronic bonding can be observed by near and extended edge x-ray absorption

(1) SXR absorption spectroscopy reveals DOS of warm dense Cu created by isochoric heating using short optical pulses





- How does the electronic DOS change upon heating to eV temperatures?
- Can we determine the electron temperature?
- How do electron heat capacity and electron-phonon coupling change?
- Can we make the measurement before the foil explodes?

Absorption spectroscopy using synchrotron pulses and streak camera detector with ps resolution probes WDM

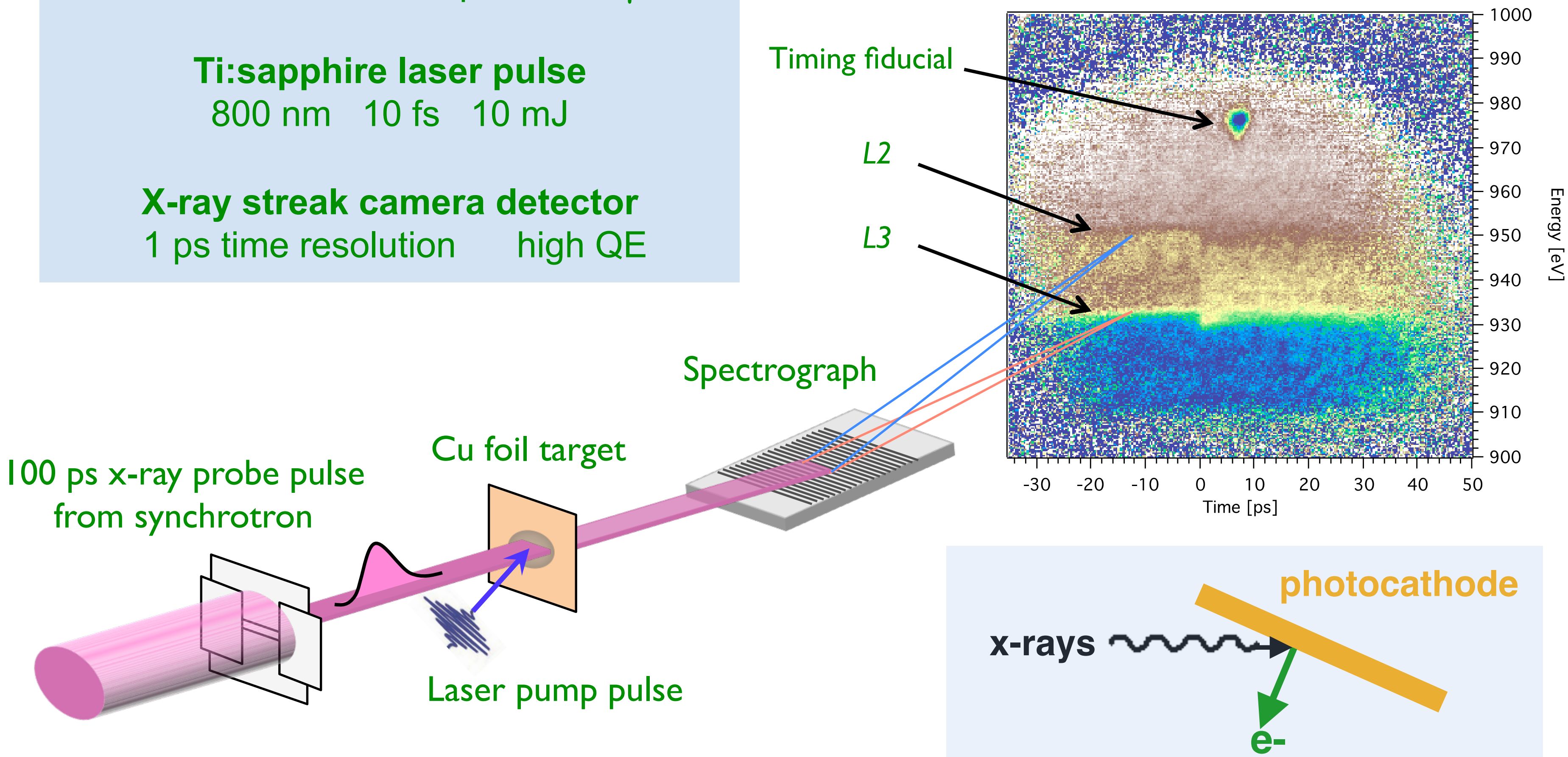
X-ray beamline at ALS synchrotron
broadband x-ray pulse
250-1500 eV   100 ps 10^6 γ

Ti:sapphire laser pulse
800 nm   10 fs   10 mJ

X-ray streak camera detector
1 ps time resolution   high QE

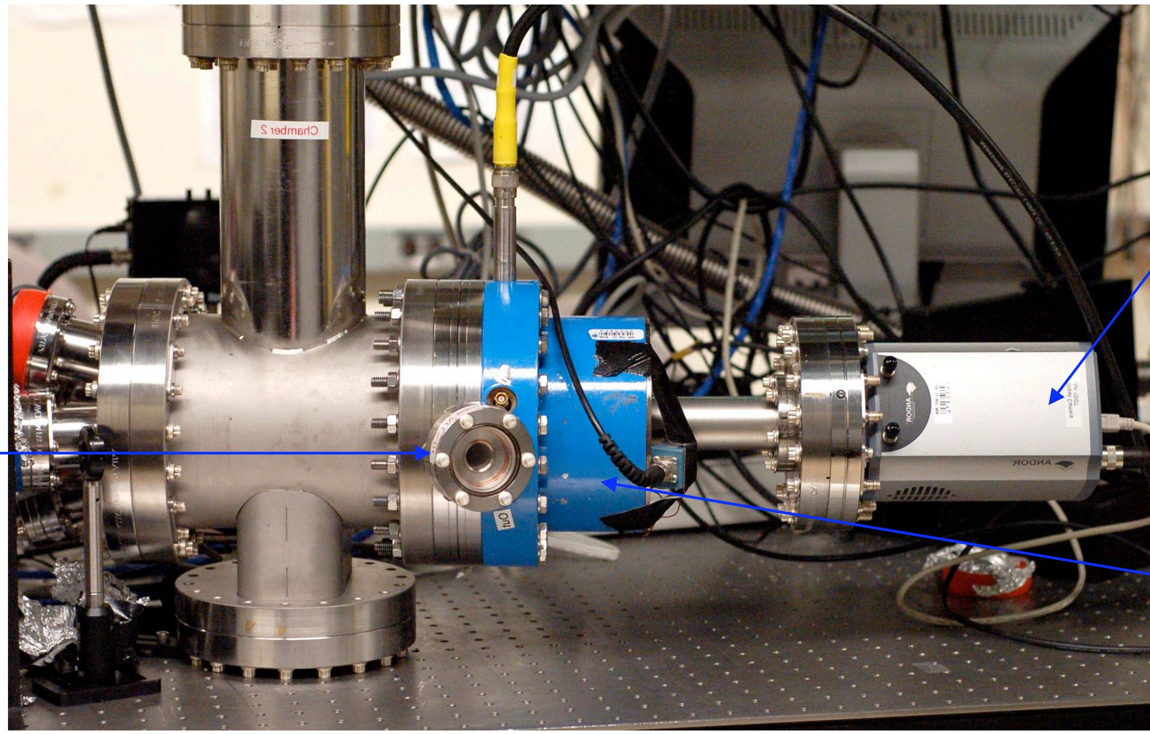
Streak camera

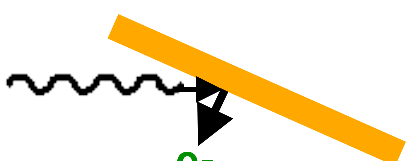
data showing Cu L-edge absorption vs. time



Feng, et al., Appl. Phys. Lett. 96, 134102 (2010)

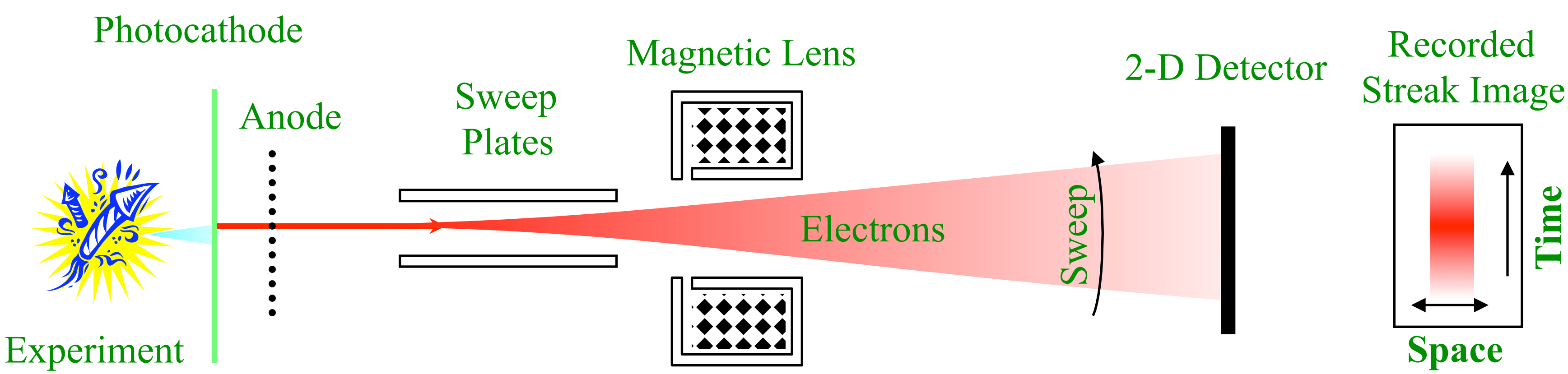
picosecond time resolution x-ray streak camera





- Improved detector sensitivity using grazing incidence
- Lowney et al., Rev. Sci. Instrum. 75, 3131 (2004)
- Feng et al., Appl. Phys. Lett. 96, 134102 (2010)

Ultrafast "x-ray streak camera"enables high-speed recording

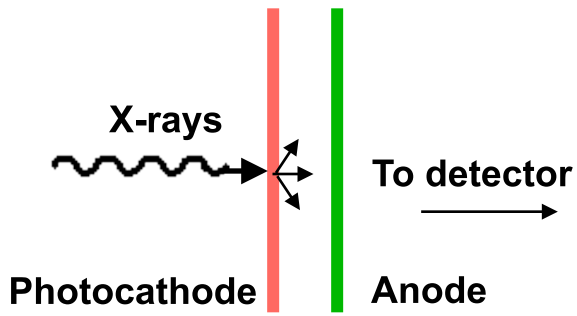


With both space and time resolution, the streak camera can record the changing spectral response of materials

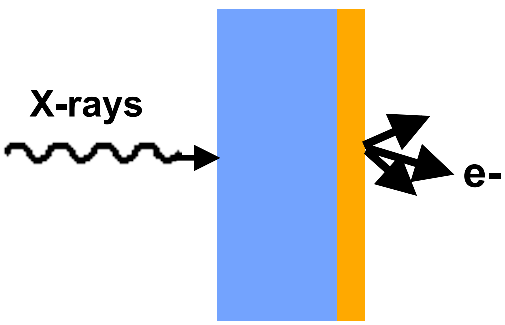
Normal Incidence vs. Grazing Incidence

- **Intensity of current picosecond/femtosecond X-ray sources implies more efficient detectors needed**

Transmission Photocathode

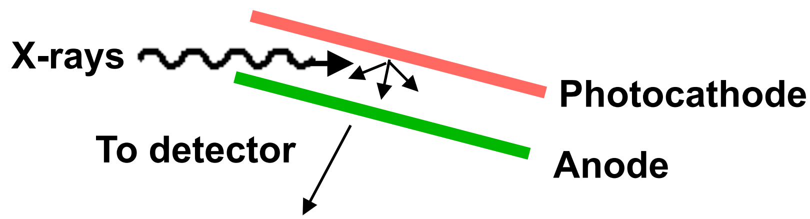


~ 0.1 — 10 %

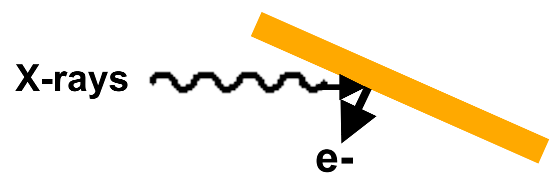


Thickness optimization

Reflection Photocathode



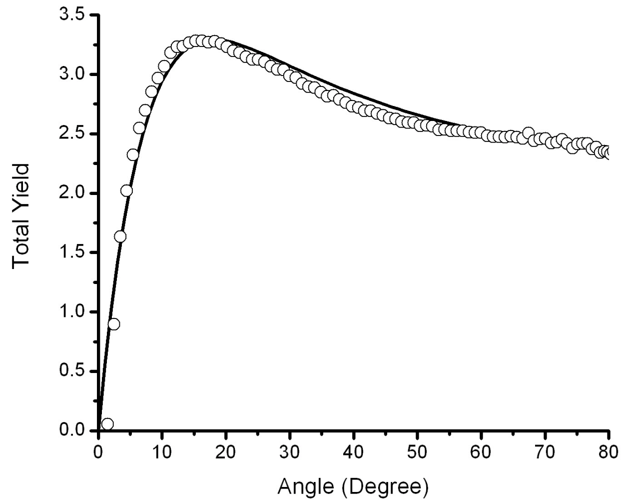
~ 100 %

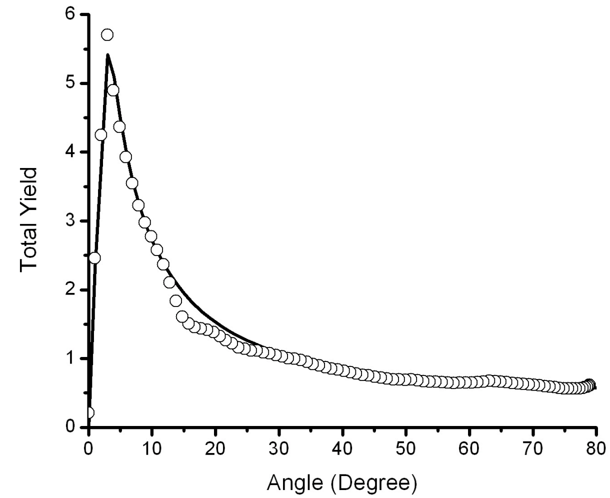


X-ray penetration and secondary electron escape depths matched.

Reflection Cathode Characterization

Total Electron Yield

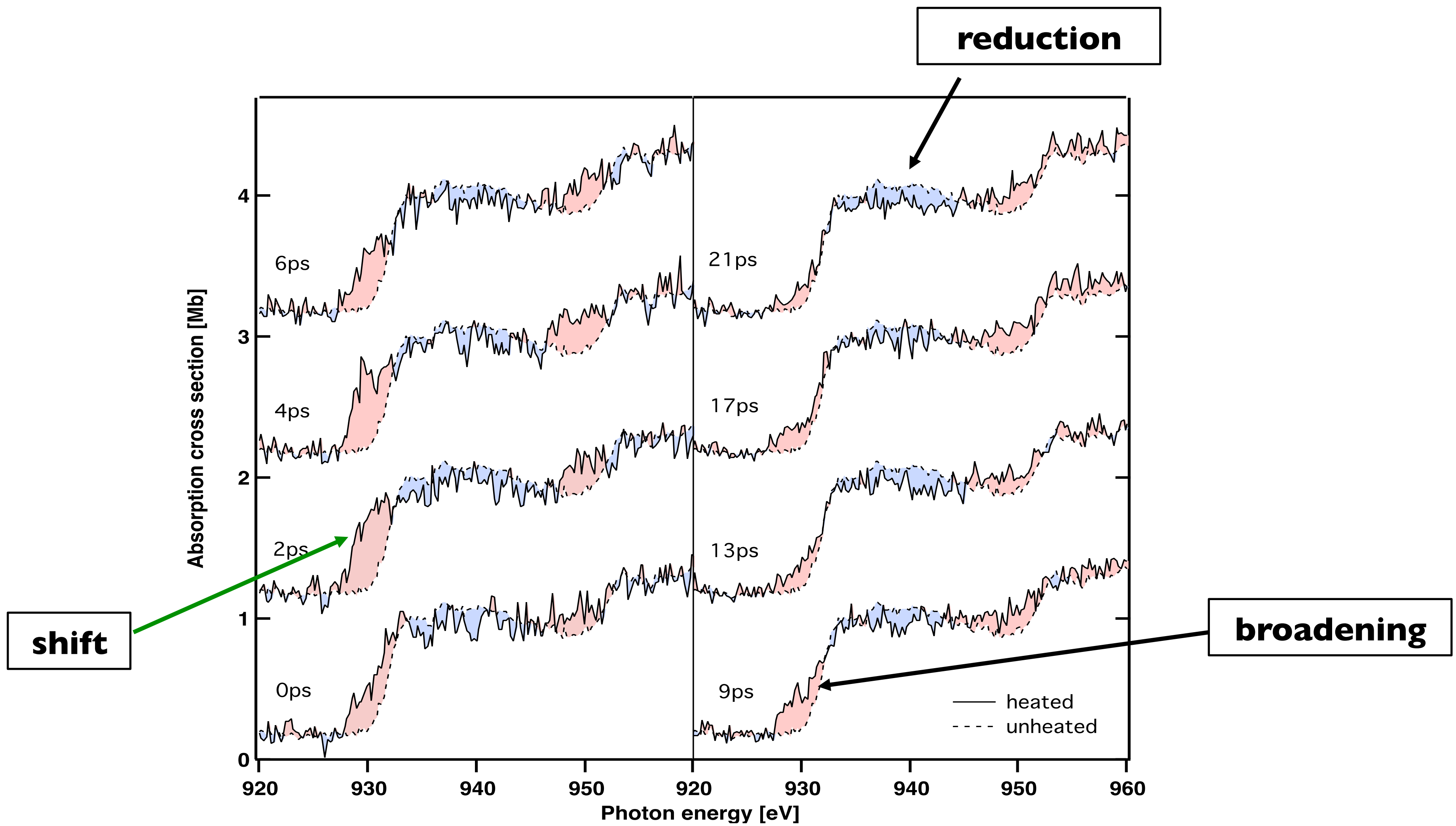




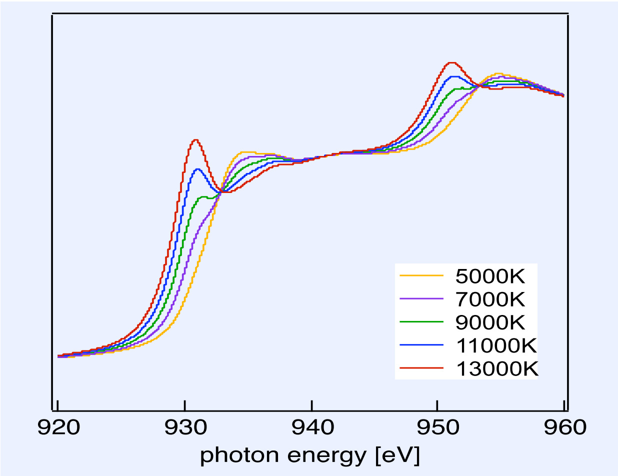
- E = 100 eV, F = 1 kV/mm
- Penentration depth of x-rays matches secondary electron escape depth.

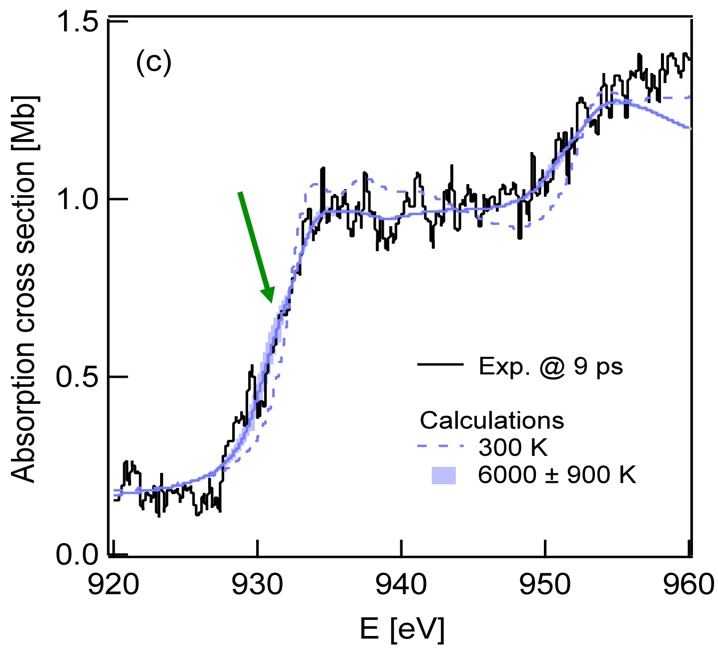
- E = 500 eV, F = 1 kV/mm
- X-ray penetration depth varies as cosecant α.

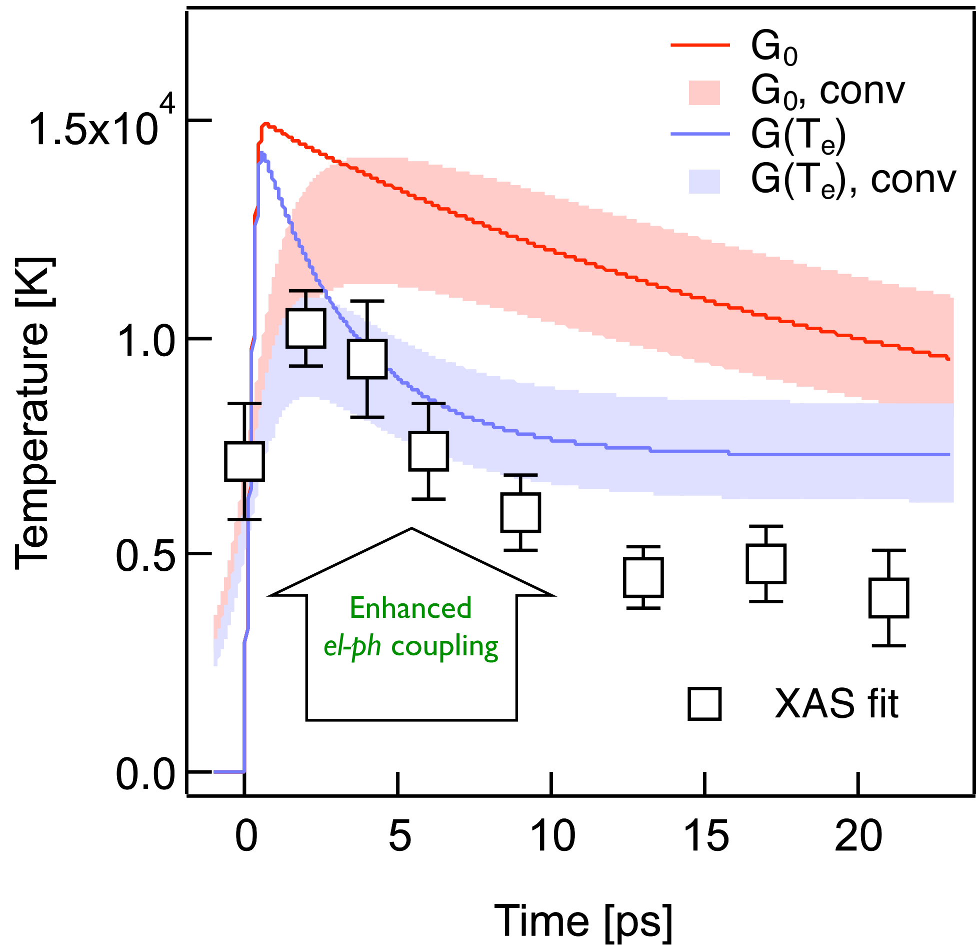
Changes in L-edge x-ray absorption in heated Cu reveals changes in DOS



Time (temperature) dependent electron-phonon coupling describes electronic structure dynamics and is compared to models



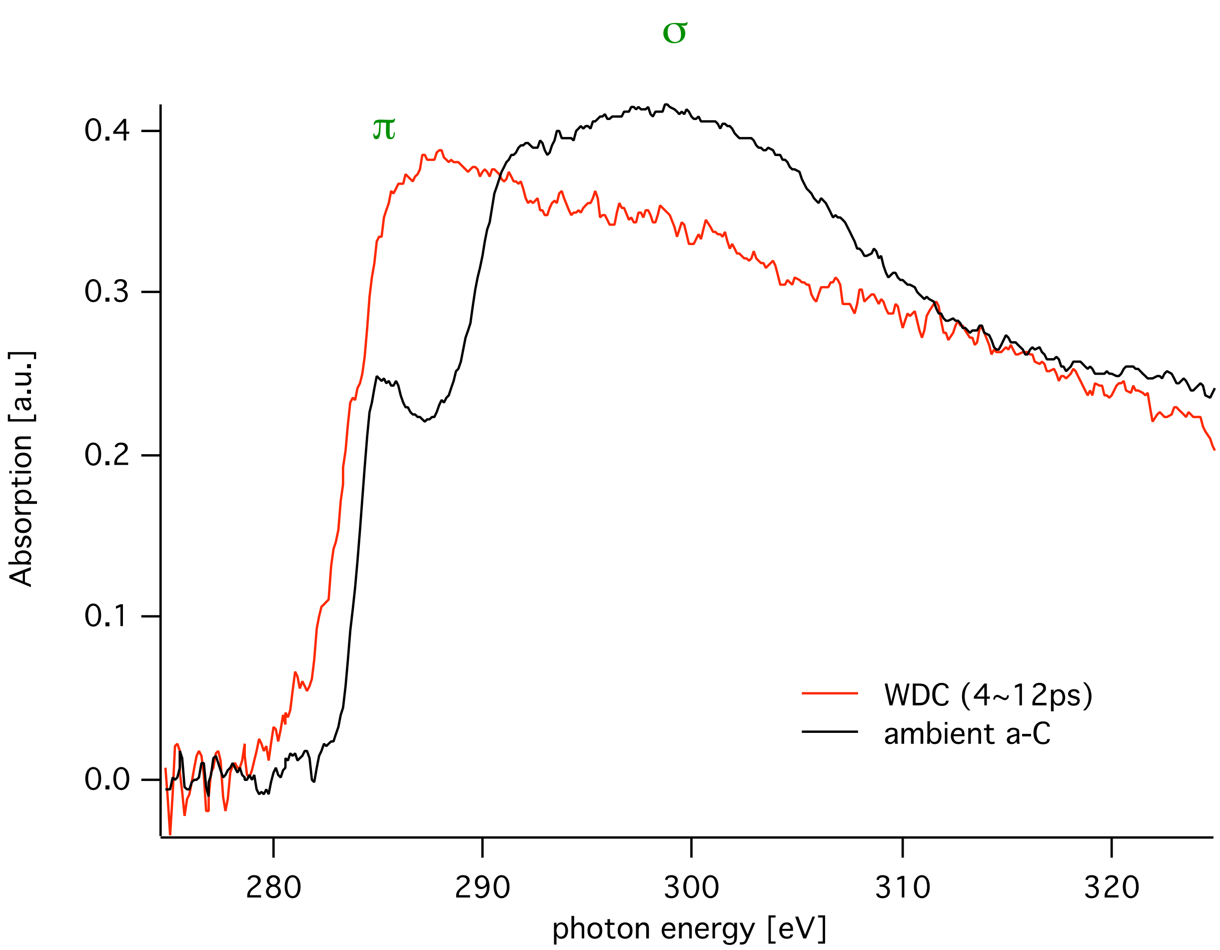


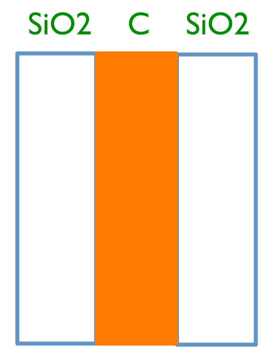


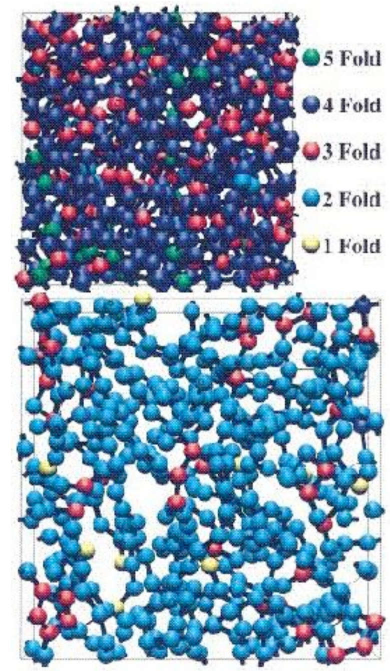
Other materials studied include C and SiO_2
Cho et al, PRL **106** 167601 (2011)

Other materials studied include heated carbon (tamped)

Tamped amorphous C
ρ=2 g/cm^3

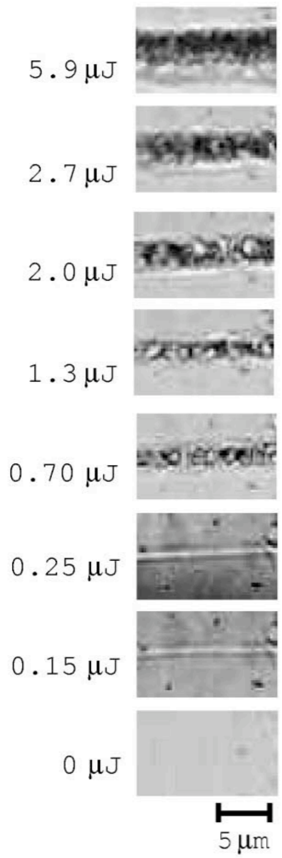




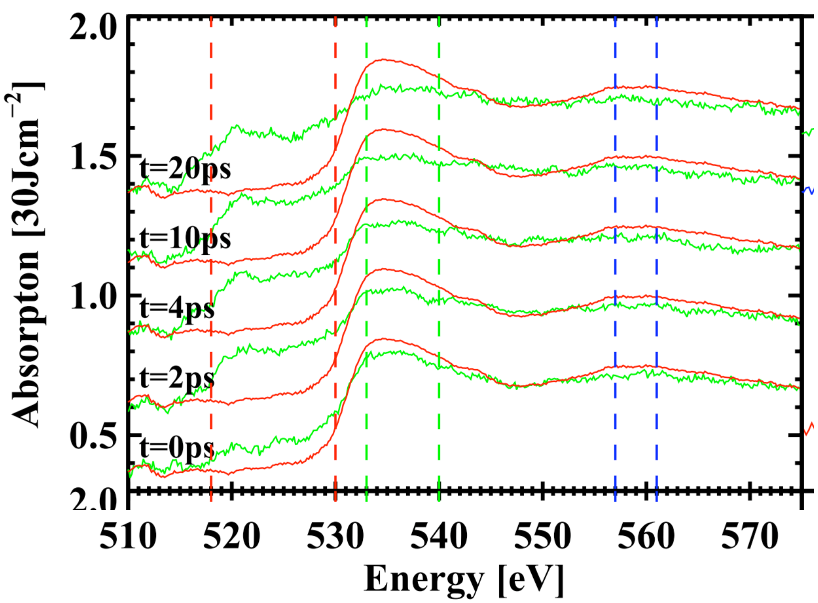


Other materials studied include "internally vaporized" SiO_2

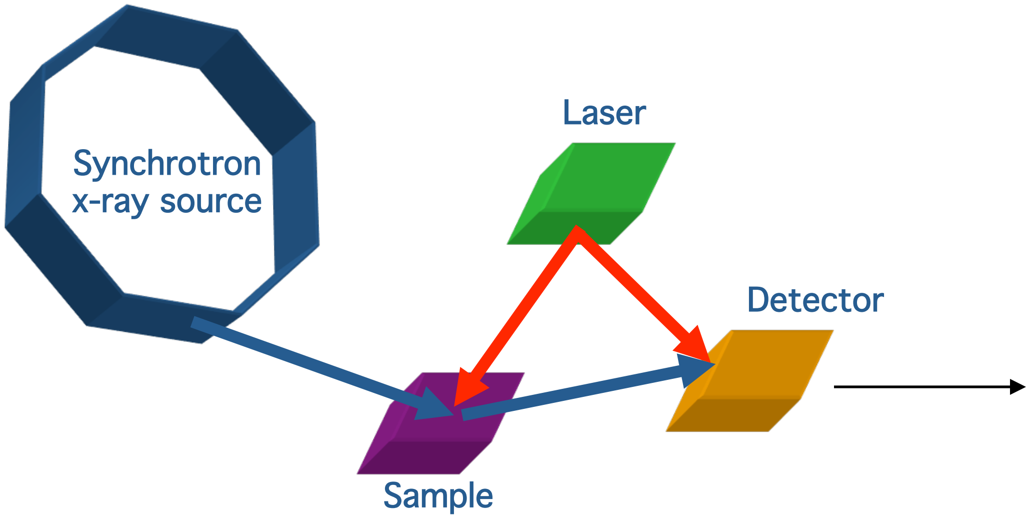
**Chan et al (Opt. Lett. 26, 1726 (2001)
used focused fs laser pulses
to write waveguides in fused silica**

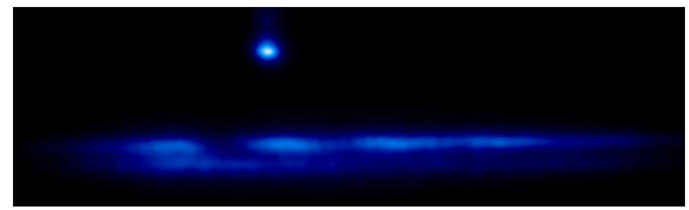


**We observed x-ray absorption of
heated fused silica at O-edge**

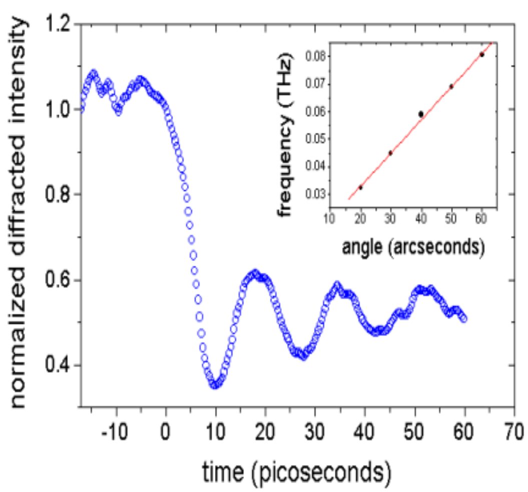


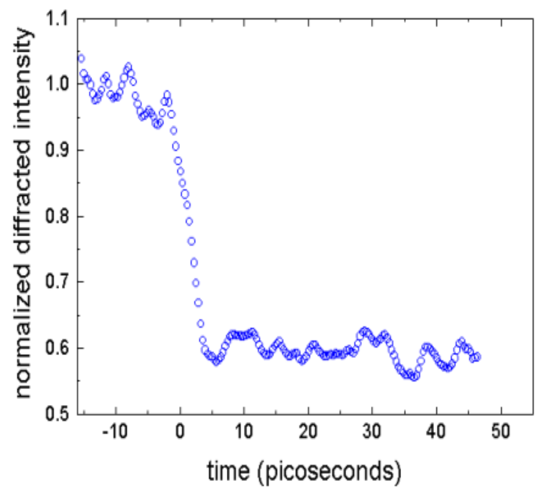
(2) Time resolved x-ray diffraction revealed structural dynamics using streak camera and hard x-rays in earlier work





Low flux conditions

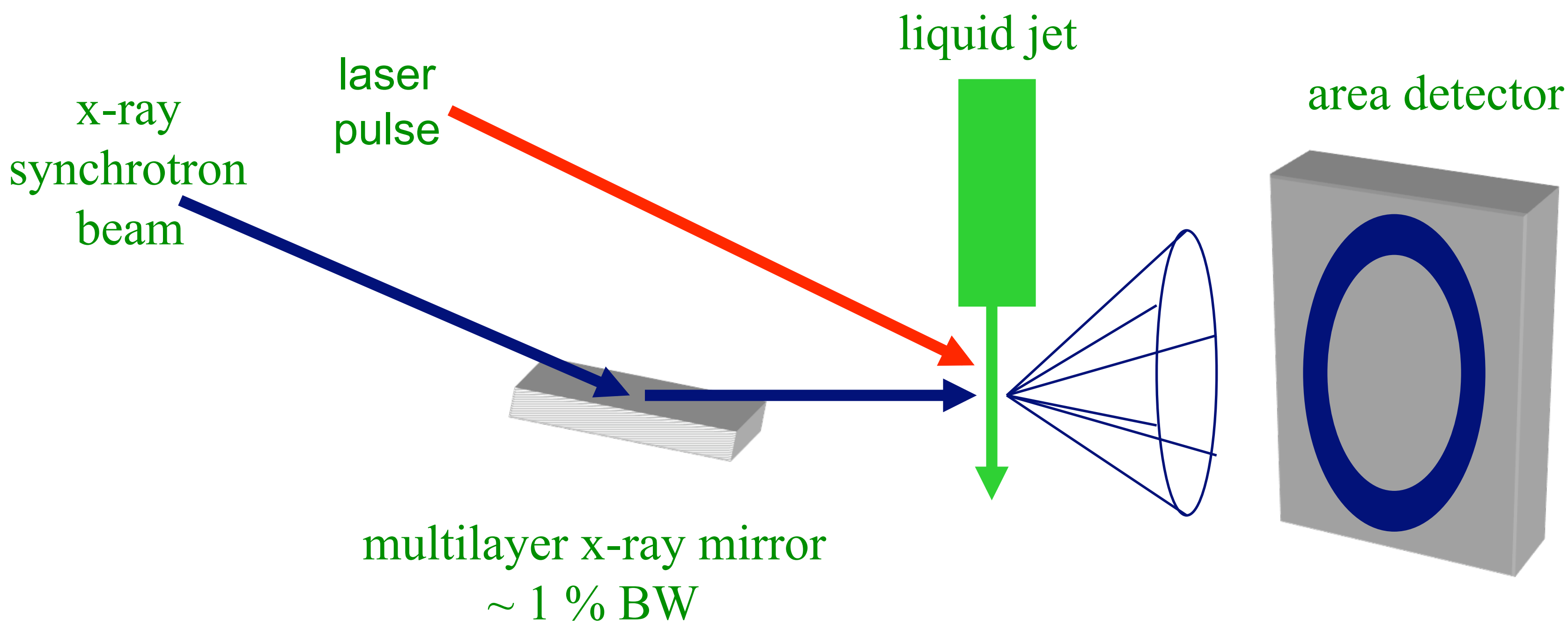




High flux conditions

Lindenberg et al., Phys Rev. Lett. 84, 111 (2000)

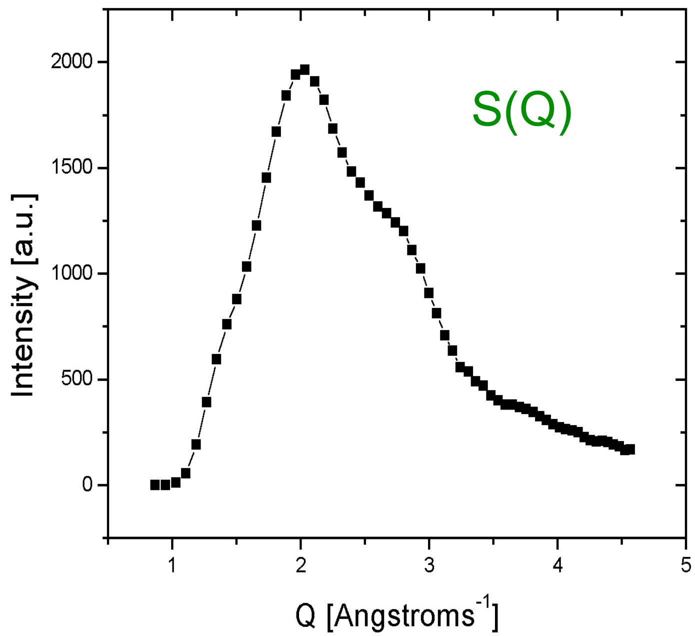
Perturbed liquid structures and dynamics can be probed by small and wide angle x-ray scattering



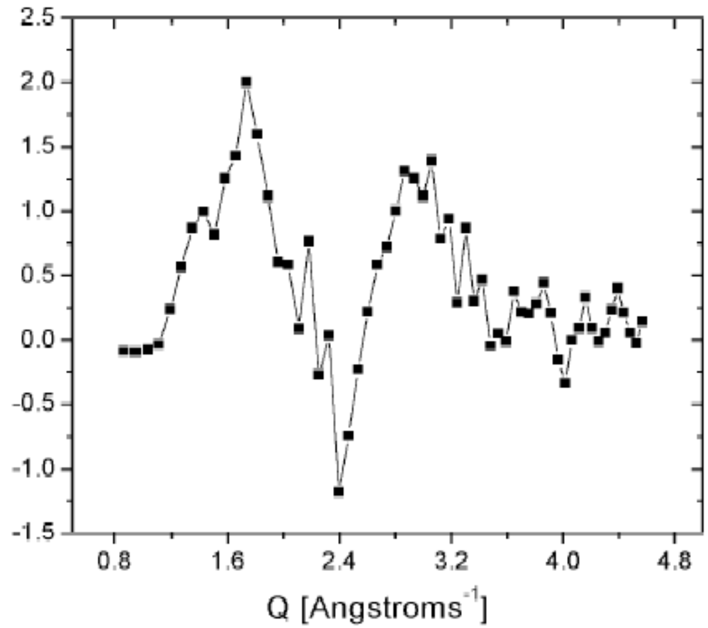
A. Lindenberg

Time-resolved structural changes in H_2O are seen upon charge injection

Static scattering signal



Difference signal at 100 ps following charge injection



A. Lindenberg

- Implies molecular re-orientation around injected charge with similarities to thermally induced changes

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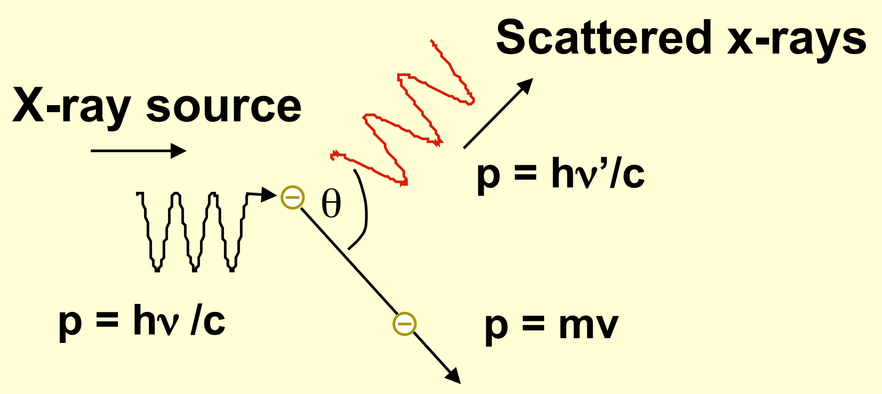
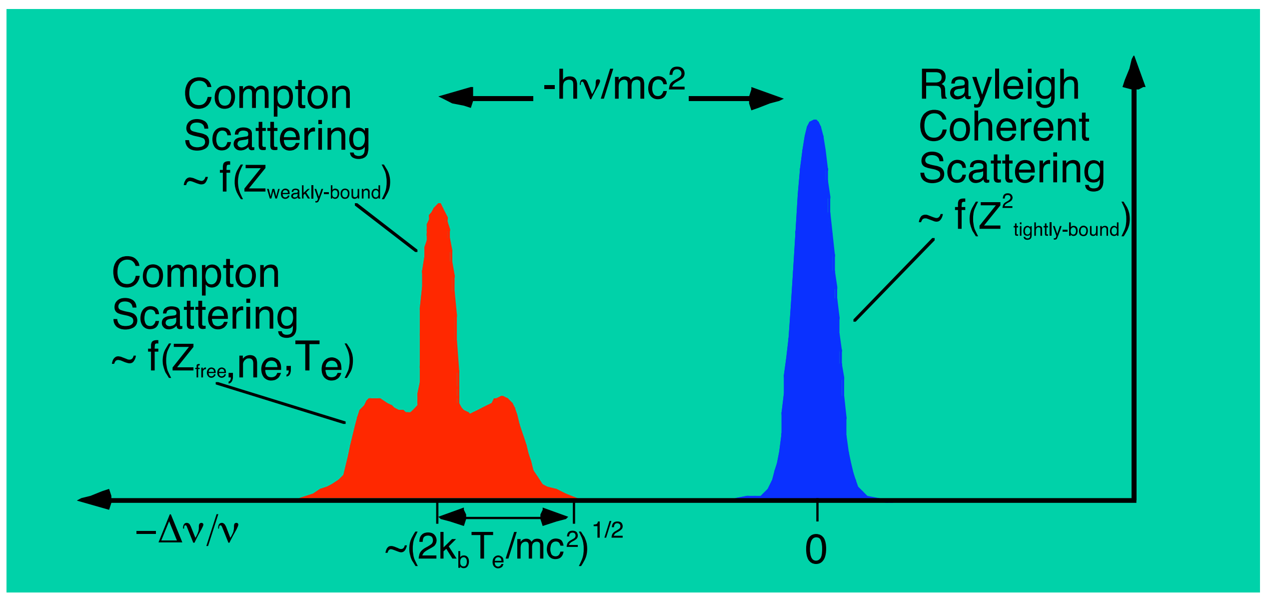
(3) Elastic and inelastic x-ray Thomson scattering from ionized material enables determination of plasma properties



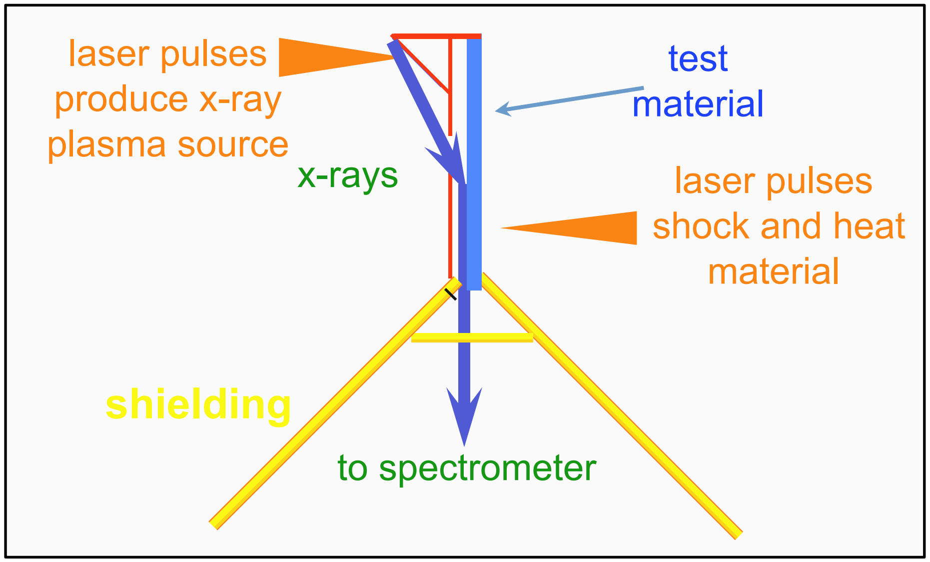
$$\lambda = \lambda_0[1 + 2(h\nu/mc^2)\sin^2(\theta/2) \pm (v/c)\sin\theta/2]$$

By varying the scattering angle, collective modes of dense matter can be probed

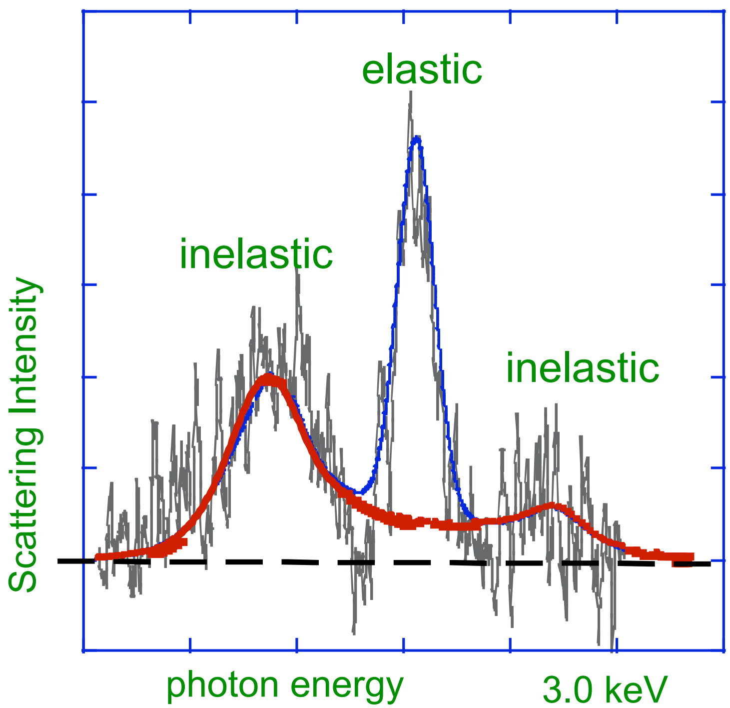
Reveals temperatures, densities, mean ionization, and velocity distribution from scattering signals (shape and ratios)



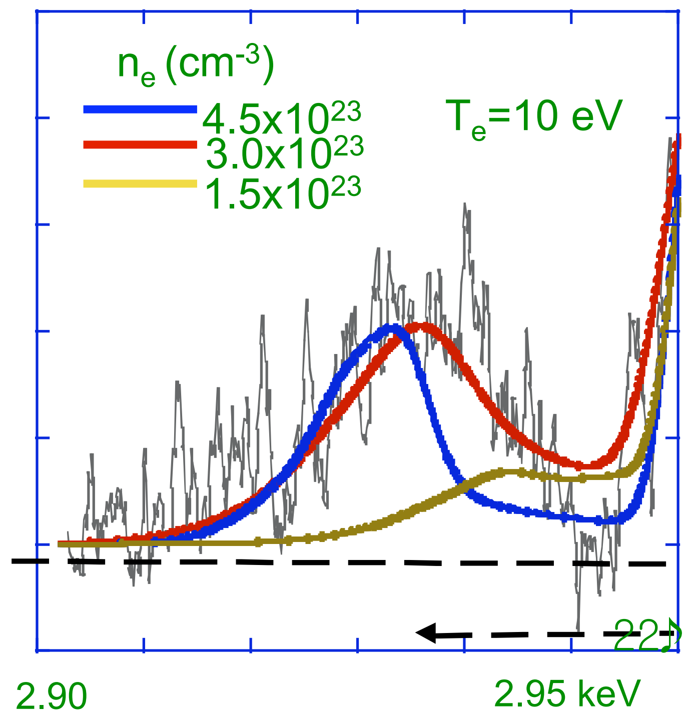
X-ray Thomson scattering can be used to probe dense plasma



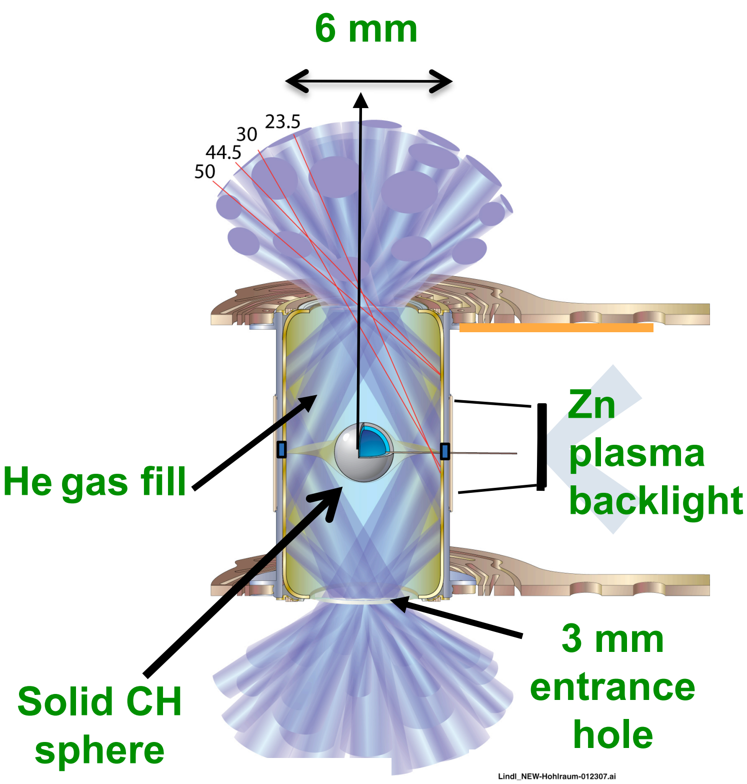
Typically need about 1 mJ of x-ray energy at 3-18 keV to probe a dense plasma using inelastic x-ray scattering (plasma backlighter or x-ray FEL)



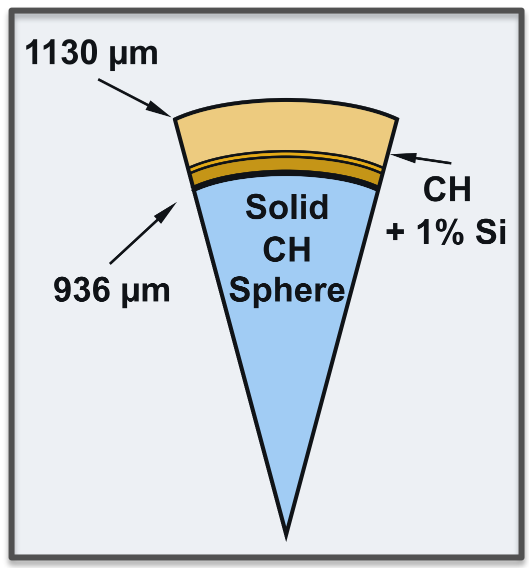
H. J. Lee, et al PRL (Be at LLE)

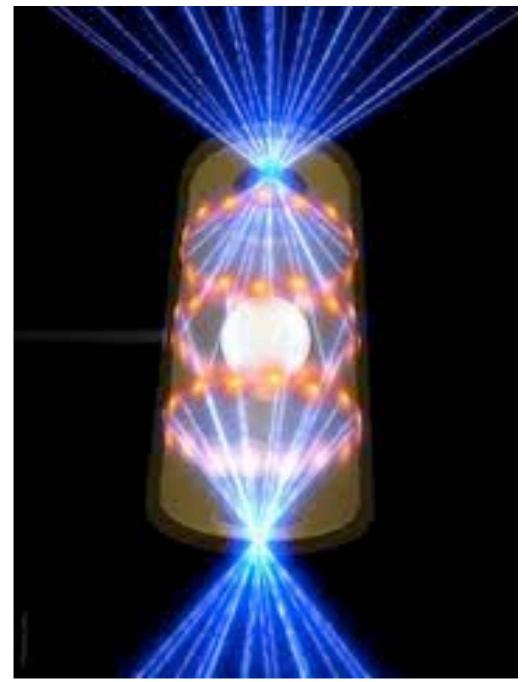


Planned experiment: Create Gbar pressures on a mm-scale sphere using NIF MJ laser facility and inertial fusion platform



Hohlraum cylinder with CH sphere in center





Simultaneous measurements at Gbar pressures: x-ray radiography and Thomson scattering

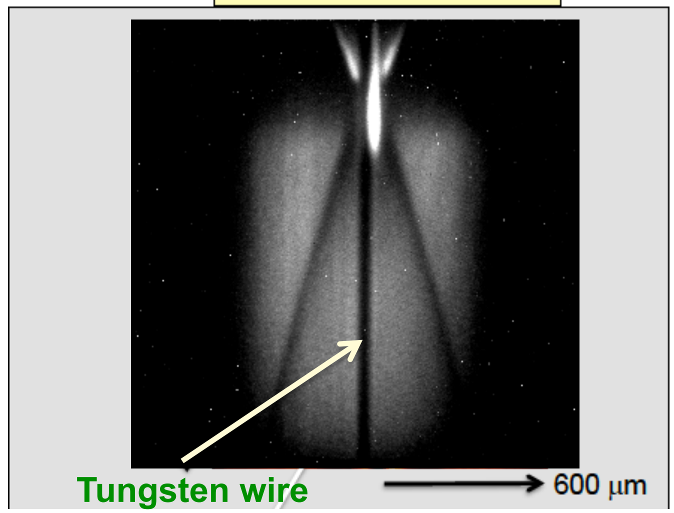
1) Measure a streaked radiographic signal from compressed matter at Gbar pressure and infer mass density profile

2) Measure an x-ray Thomson scattering signal from compressed matter at Gbar pressure and infer electron temperature

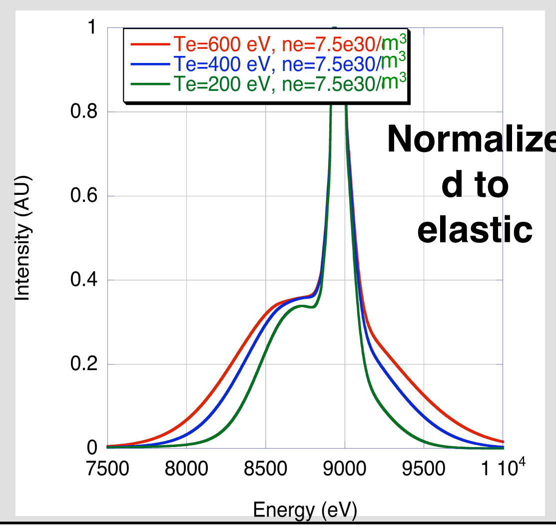
3) Use this information to constrain the EOS of matter at Gbar pressure, created using MJ laser driver pulse

Collaboration among
UC Berkeley, GSI, Germany; SLAC;
U Jena, U Rostock; UCLA; Imperial College; Carnegie Institute

Composite "streak"



X-ray Thomson Scattering



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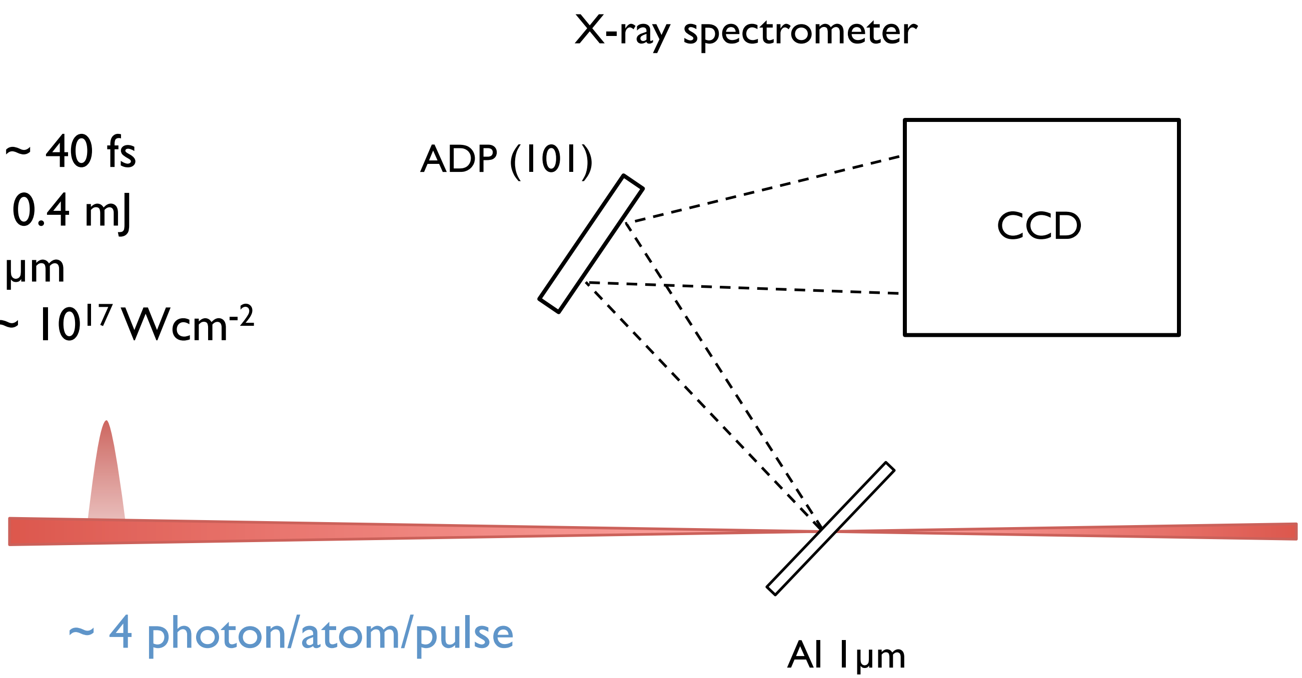
X-ray emission spectroscopy for X-ray heated Aluminum

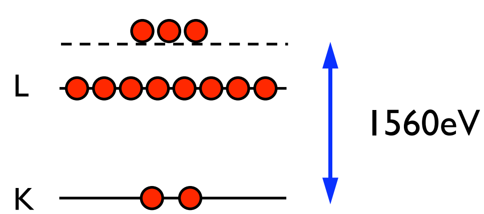
X-ray parameters

$\hbar\omega$: 1480 ~ 1800 eV
bw ~ 0.5 %

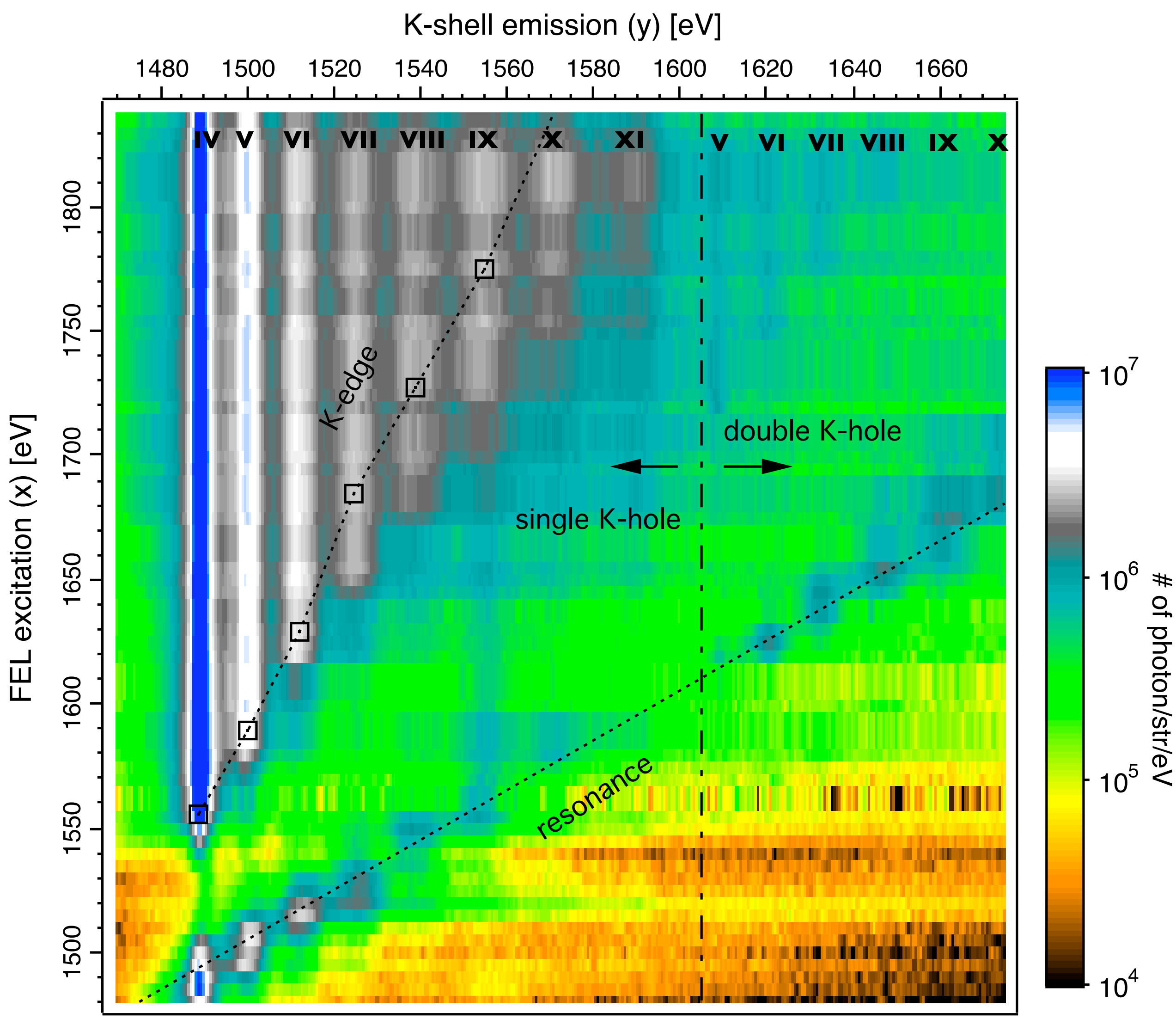
Pulse duration ~ 40 fs
Pulse energy ~ 0.4 mJ
Focal spot ~ 3 μm
Peak intensity ~ 10^{17} Wcm^{-2}



With Justin Wark's group at Oxford and scientists at LCLS



K-shell emission patterns indicate various ionization processes



Laser heated materials are dynamic

- energy deposition of ~ one quanta per unit cell drives structural and other property changes that can be probed… by time-resolved x-ray scattering

- a variety of quanta can be utilized to “heat” or “vaporize” materials
  - THz to far-IR drives phonon modes
  - near-IR excites electrons from valence to conduction bands
  - optical to uv excites electronic transitions and charge transfer states
  - soft x-rays couple core levels to valence states
  - hard x-rays penetrate and excite larger volumes

- coupling of excitation to various modes is defined by the time-scale
  - time << picoseconds can involve non-thermal processes
    - photochemistry, electron re-scattering
  - time >> picoseconds involve thermal processes
    - mode diffusion, ablation
  - time = scale length <u>divided by</u> relevant velocity

Laser heated materials can be studied by pump-probe techniques at a variety of facilities

- small-scale laboratories provide intense, short-pulse lasers to create and probe warm (high-energy-density) materials
  - probes include plasma x-ray sources, high-harmonic sources

- intermediate-scale facilities include petawatt lasers, pulsed particle beams, x-ray synchrotrons, free-electron lasers, etc., and are widely accessible

- large-scale facilities allow large volume studies to extreme high-energy-density conditions, but have limited access
  - NIF megajoule laser, Vulcan PW laser, OMEGA kJ laser, pulsed power

Multiple x-ray probes at ALS and LBNL can be generated

Synchrotron - based

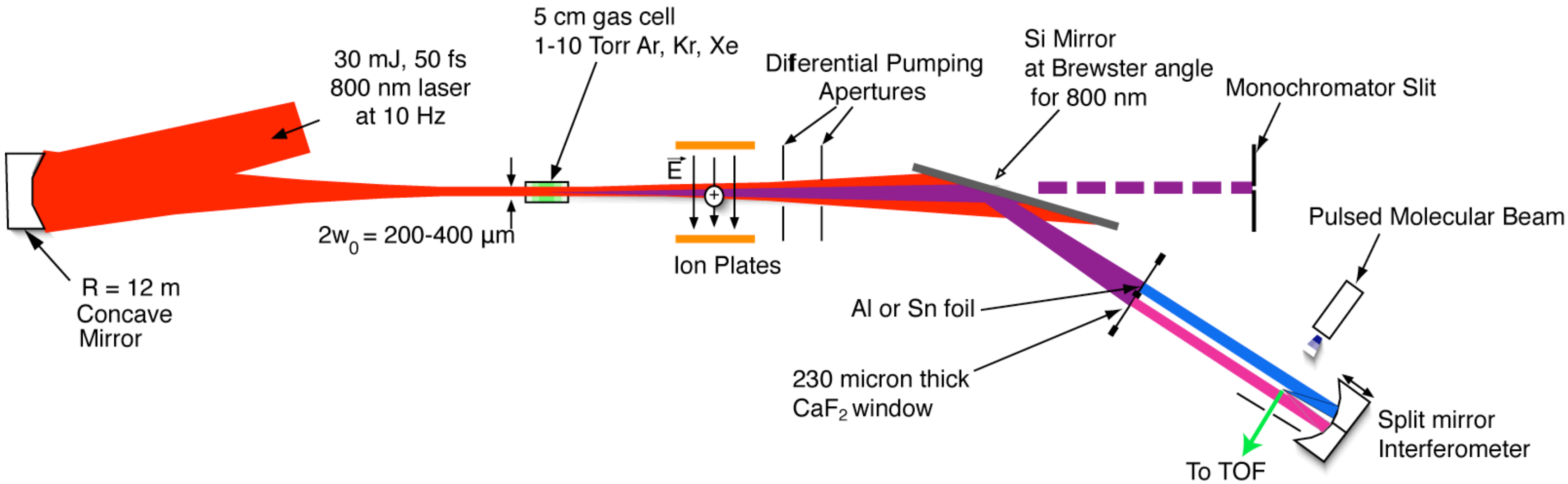
- Synchrotron light with typically 10^6 photons and 100 ps pulses
- Streak camera can give 1 ps temporal resolution
- ALS BL6 covers 250 - 1500 eV, with monochromatic x-rays at higher energy (to 12 keV)
- X-ray absorption measurements of isochoric heated samples done at C & O K-edges, and Ni & Cu L-edges

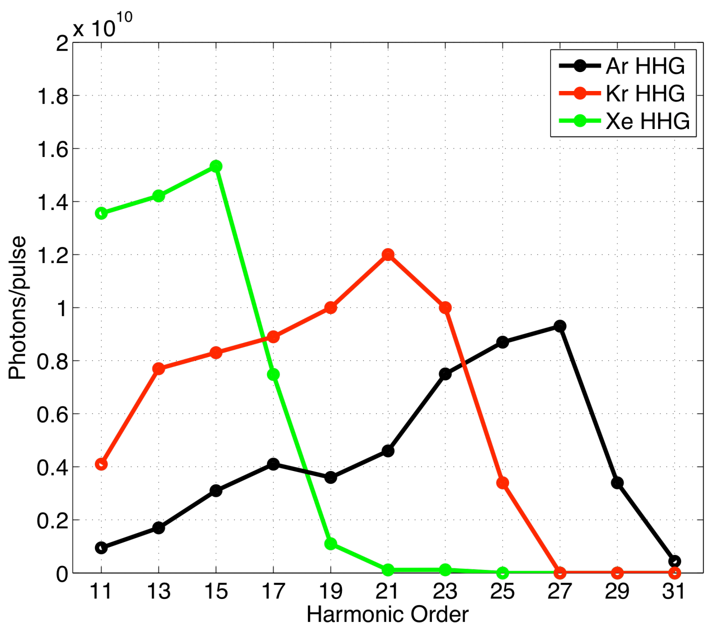
- Sliced x-ray pulses with ~ 10^2 photons per pulse at 200 fs

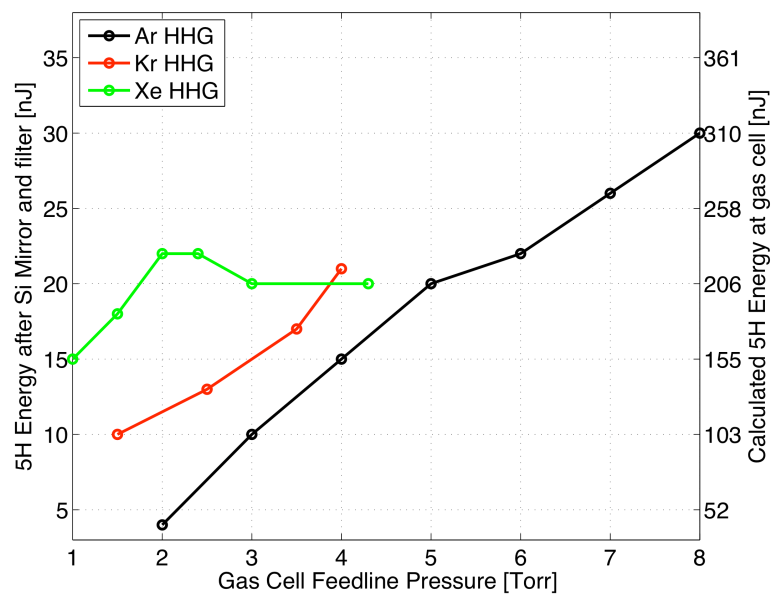
Laser - based

- Plasma backlighters with ~ 10^{-6} efficiency into narrow angle
- High laser harmonics create with ~ 10^{-6} efficiency in SXR

Soft x-rays can be generated on a table top using harmonic generation in gases





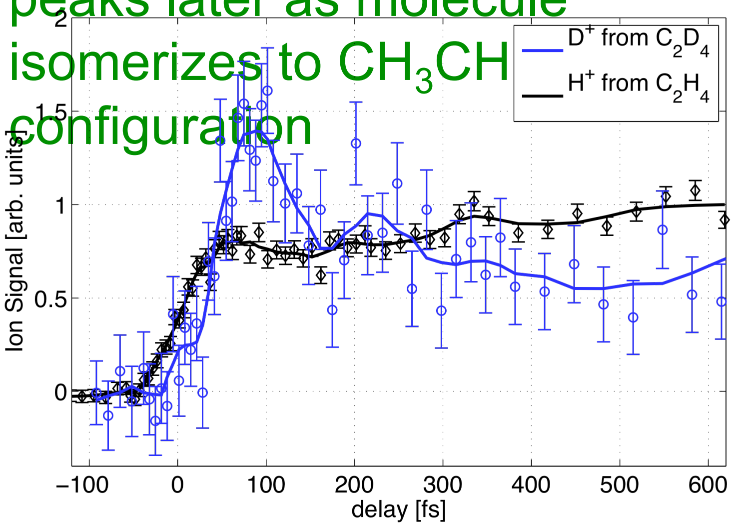


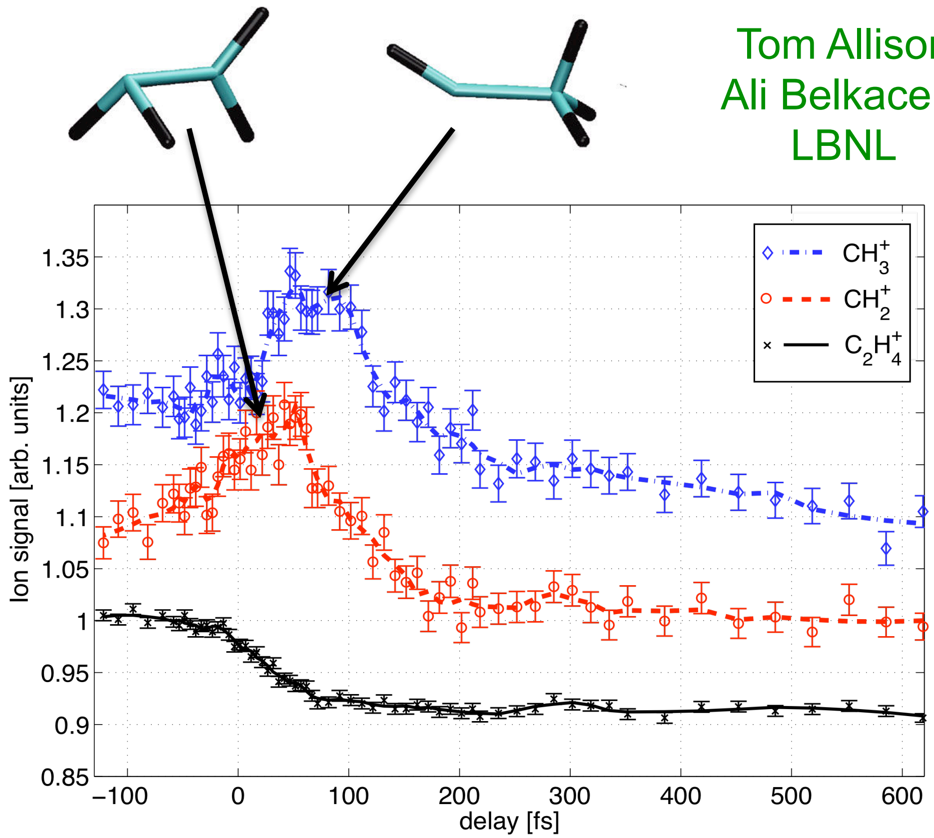
VUV pump and XUV probe time scale of C - C bond breakage

$C_2H_4^+$ signal quickly decays as system moves from Franck-Condon Region

Symmetric breakup (CH_2 + CH_2^+) peaks on time scales comparable to electronic relaxation

Asymmetric breakup ($CH+CH_3^+$) peaks later as molecule isomerizes to CH_3CH configuration





Tom Allison
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LBNL

Consistent with electronic relaxation dominated by CH_2CH_2 Conical Intersection

The Berkeley campus program (funding - SSAA to Joint Program) utilizes tools in Bay Area for studies of WDM, HDM, and plasmas

ALS provides short-pulse lasers for isochoric heating of matter and broad-spectrum synchrotron backlighters, and detectors with sufficient time resolution for rapid absorption measurements of near and extended edges for electronic structure measurements.

NIF and Jupiter lasers provide drivers for shock compressing matter and backlighters with sufficient photon energy and flux for inelastic scattering measurements of electron & ion temperature & densities and radiography.

LCLS provides x-ray pulses with sufficient energy for isochoric heating of matter, with sufficient photon energy and flux for inelastic scattering measurements of electron & ion temperature & densities, as well as high peak power and high energy optical lasers, to drive plasmas and shock compressed matter.

NDCX can similarly create relevant conditions for appropriate probing.